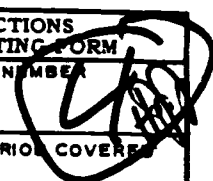



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## FINAL REPORT (GFR)

Title: "Theoretical Studies Relating to the Interaction of Radiation with Matter"

Supported by: The U.S. Office of Naval Research

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The final report for Grant No. N00014-91-J-1146 is divided into seven subject areas as follows:

1. Coherent transients using broadband optical pulses
2. Four-wave mixing in magnetically degenerate systems
3. Interpretation of laser cooling mechanisms
4. Laser cooling in one and two dimensions
5. Nonlinear spectroscopy of cold atoms
6. Transient spectroscopy using counterpropagating pulses
7. Theory of four-wave mixing

A detailed description of the work carried out under this Grant can be found in the articles listed at the end of this report as well as in Annual Reports GAR1 and GAR2 associated with this Grant. In this Final Report, the research areas are summarized briefly, with references to the appropriate articles.

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## 1. Coherent transients using broadband optical pulses

We have studied the interaction of two broadband optical pulses with an ensemble of three-level atoms.<sup>8\*,12,16,18</sup> The two pulses are time-delayed by an amount  $t_{12}$  relative to one another and the atomic response to these pulses is probed by a third pulse. The signal, monitored as a function of  $t_{12}$ , is found to exhibit a sharp peak or dip centered at  $t_{12}=0$ , whose width is *narrower* than the pulse correlation time. This phenomenon is explained in terms of the so-called dark resonances. For  $t_{12}=0$ , a linear combination of the atomic states is decoupled from the fields; for  $t_{12}\neq 0$ , the decoupling is not complete. The resultant signal width scales as the pulse correlation time divided by a dimensionless saturation parameter. The effect can serve as the basis for spatially selecting atoms in a given internal state. Experimental confirmation of the theory has been reported by LeGouet's group in Orsay, France.

## 2. Four-wave mixing in magnetically degenerate systems

We have concluded the analysis of a four-wave mixing experiment carried out in D. Steel's laboratory at the University of Michigan. In that experiment, the four-wave mixing signal was studied as a function of the detuning between two of the applied fields. A narrow (width less than the excited state width) structure appears in the line shape which can either grow or diminish as a perturber gas is added to the sample. Whether or not the narrow signal grows is linked to the polarizations of the applied fields. The authors could not explain why the narrow resonance disappeared with increasing perturber pressure for parallel-polarized fields. We have analyzed this problem, taking into account the multi-level structure of sodium [G. Rogers, PhD thesis, New York University, 1992]. Our results give a consistent explanation of the experimental results of Steel and coworkers. The narrow resonance is seen to disappear for parallel polarization as a result of pressure broadening of the optical transition and collisionally-induced mixing of the excited state sublevels. Redistribution of atomic velocities owing to velocity-changing collisions also plays a role. The of velocity selective optical pumping processes which contribute to the line shape are compared with those which enter in theories of sub-Doppler laser cooling.

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\*Superscripts refer to the numbers in the list of publications found at the end of this report.

### 3. Interpretation of laser cooling mechanisms

We have given a consistent picture of laser cooling in *both* the Doppler and sub-Doppler limits based on the matter gratings formed by the incident fields.<sup>19</sup> Using results from nonlinear spectroscopy, we are able to explain in a unified manner the various features encountered in laser cooling. In particular, the switchover from cooling to heating in an intense standing wave field and the sub-Doppler cooling in multi-level atoms are found to have direct analogues in the pump-probe spectroscopy of atomic systems. Laser cooling is also analyzed in terms of its effect on the incident fields. It is shown that the friction force of laser cooling can be related to the absorption of each field and the back-scattering of each field off the matter gratings produced by the fields. The back-scattering also leads to an exchange of energy between the fields. Using this model, it is easy to understand why, for parallel polarizations of the incident fields, narrow resonances can appear in nonlinear spectroscopy even though the same field geometry does not lead to sub-Doppler cooling.

### 4. Laser cooling in one and two dimensions

We studied laser cooling in one dimension produced by two, counterpropagating, linearly polarized fields whose polarization angles differ by an amount  $\theta$ .<sup>13</sup> Since sub-Doppler cooling is known to exist for  $\theta=\pi/2$  and not to exist for  $\theta=0$ , one might think that there is a monotonic increase in the friction force of laser cooling as  $\theta$  varies from 0 to  $\pi/2$ . We have found this *not* to be the case. Instead, for small angles  $\theta \ll 1$ , the friction force can actually be larger than that for  $\theta=\pi/2$ . The result is linked to the fact that there is a rapid variation of the atomic population difference density in the vicinity of the quasi-field nodes which occurs for  $\theta \ll 1$ . A similar feature appears in the two-dimensional cooling of an atomic vapor by two, orthogonal, standing wave fields which are linearly polarized in orthogonal directions. In the 2-D case, the situation is somewhat more dramatic as the friction force actually has a logarithmic dependence on velocity for small velocity.<sup>17</sup> Our calculations show that "standard" methods for calculating the friction coefficient are doomed to failure for the limiting cases we studied, owing to the rapid variation of population density near the field nodes. Diffusion and particle localization have also been studied for the 1-D case and channeling of the particles in momentum space has been studied for the 2-D case.

It was found that the depth of the optical potentials was sufficiently large to allow for localization of the atoms in the minima of the potentials. Thus, our calculations, in which all effects related to atomic localization are ignored, can be questioned. To evaluate the role played by atomic localization, we repeated the calculation using a fully quantized approach. We found that the qualitative conclusions reached for the sub-Doppler cooling on a  $J=1/2 \rightarrow 1/2$  transition did not change.<sup>22</sup> Thus, there is a range of laser field strengths where a lower temperature is reached if one uses an angle  $\theta \neq \pi/2$  between the two field polarization vectors. The distribution of populations in the various bands of the optical potential was also studied as a function of  $\theta$ .

### 5. Nonlinear spectroscopy of cold atoms

Motivated by recent experiments of Kimble's group<sup>2</sup> and the laser cooling group in Paris,<sup>3</sup> we began a study of the pump-probe spectroscopy and four-wave mixing of cold atoms. For an ensemble of two-level atoms, we predict the existence of a new class of resonances whose origin can be traced to the photon recoil effect. These *recoil-induced resonances* (RIR) appear in the sub-Doppler limit and have a shape which mirrors that of the atomic velocity distribution.<sup>14</sup> As such, nonlinear spectroscopy can be used as a probe of the velocity distributions achieved by laser cooling. The calculation was extended to include effects related to the magnetic degeneracy of the atomic levels, a feature critical to sub-Doppler cooling. We found that the RIR persist even in the presence of optical pumping,<sup>21</sup> although they may be obscured in some cases by effects related to atomic localization. The narrow structures seen recently in pump-probe spectroscopy of cold atoms may provide experimental confirmation of these resonances.

### 6. Transient spectroscopy using counterpropagating pulses

We have studied the interaction of a series of counterpropagating pulses with an atomic ensemble of cold or thermal atoms.<sup>16</sup> In a scheme we refer to as a grating stimulated echo (GSE), the atoms interact sequentially with two counterpropagating pulses, a standing wave pulse and a third traveling wave pulse. The first two pulses create a spatially modified ground state population which dephases as a result of inhomogeneous

broadening. The standing wave pulse reverses the direction of the dephasing so that the populations can rephase at some appropriate time after the application of the standing wave pulse. The rephasing is probed by the third traveling wave pulse which leads to an echo signal. The GSE is a very sensitive probe of any velocity changes experienced by the ground state atoms. As such it has potential applications as a probe of collisional processes, a probe of photon recoil effects, and as an accelerometer. Calculations along these lines are in progress. We have also considered a variant of the GSE in which the time delay between the first two pulses is set equal to zero. We have solved this problem in a perturbation theory limit for arbitrary polarizations of the applied fields and arbitrary fine and hyperfine structure. The resulting magnetic grating echo (MGE) signal can be used to determine the various hyperfine splittings. Articles on these results are in preparation.

## 7. Theory of four-wave mixing

A theory of four-wave mixing was formulated using an *amplitude* approach in the Schrodinger picture.<sup>20</sup> The calculation properly accounts for the repopulation of the ground state of the "two-level" atoms resulting from spontaneous emission. Until this calculation, no one has been successful in developing such a theory. The interplay between the excitation and emission at the various atomic sites is evident in this amplitude approach, which involves fourteen distinct contributions to the final state amplitude. The technique is extended to include effects related to the recoil-induced resonances mentioned above, as well as the so-called pressure induced extra resonances. Surprisingly, the amplitude calculation is actually simpler than the density matrix calculation in certain limits, as the final state probability can be rewritten in an especially simple form.

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